Correlation between Stick - Slip events and contact charging in the dynamic friction at nano-scales

G. Ananthakrishna

Materials Research Centre
Indian Institute Science
Bangalore - 560012

Collaborator
Jagadish Kumar

Outline of the talk

- Introduction

- Experimental results of Putterman (PRL, 2000) - unexplained,
  Conclusions - intriguing

  - SFA on PMMA: Correlation between stick-slip events and charge transfer. Ascribe friction to bonds formed due to contact electrification.

- Modelling - Specific issues. Contact mechanics, visco-elasticity, visco-plasticity, contact electrification.

- Results.

- Summary and Conclusion.
Leonardo da Vinci, a self taught genius (April 15th 1452 – May 1519)


Sketches from the Codex Atlanticus and the Codex Arundel.
(a) The force of friction between horizontal and inclined plane
(b) The influence of apparent contact area upon the force of friction.
(c) The force of friction on a horizontal plane by means of a pulley.
(d) The friction torque on a roller an half bearing.
Anatomist

Muscles of the arm and shoulder in rotated views, c. 1510

Anatomical Studies, folio 141v

vertebral column, c. 1510,

Anatomical Studies, folio 139v

Fetus within the womb,

c. 1510-12

Anatomical Studies, folio 198r
Mechanical Engineer

Rotary ball bearing
Codex Madrid I
folio 20v

water-powered rolling mills
Codex Atlanticus
folio 10r

Two-wheeled hoist
Codex Atlanticus
folio 30v
Aerodynamics Engineer

Design for flying machine
Ms, B, folio 74v

Working model of a flying machine
Museum of history of sciences, Florence
Scientist: Turbulence

Turbulent wakes behind a rectangular plank

c. 1509-11, Windsor, Collection
Map of the Chiana valley
1504, Windsor Collection

Design for Centralized "Tample,"
c. 1488, Ms. Ashburnham I, folio 5v
Friction is the energy dissipated when two surfaces are in relative motion.

It arises due to different fundamental causes:
- Mechanical in nature: Interlocking of asperities at the sliding interface.
- Chemical in nature: Adhesion between two solid surfaces in contact.

Earlier lubricants had been used to reduce friction between sledges used for transportation.

Painting from the tomb of Tehuti-Hetep, El-Bershed (1880 BC). The colossus is secured on a sledge but there is no roller or levers.
**Introduction:**

Friction - scale dependent - macro, micro, nano scales.

*Friction and Stick-Slip:* Leonardo da Vinci, 16th century
Frictional resistance $f \propto$ load, $\rightarrow$ is independent of area $S$

Amonton 1699, Coulomb 1781: Amonton – Coulomb laws:
Minimum force required to move

$f_s = $ pulling force, $F = $ normal force, $S = $ area,
$\mu_s = $ static friction coefficient. $\rightarrow f_s = \mu_s F$

For constant velocity $v$, $f_d = \mu_d F$

$\mu_d =$ dynamic friction coefficient. $\mu_s$ and $\mu_d$ independent of $S$ and $F$. 

![Friction and Stick-Slip Diagram](image)
Introduction:

Bowden Tabor (1950):

\[ F = \sigma_y S_{eff} \]

\[ f_s = \sigma_s S_{eff} \]

\( \sigma_s < \sigma_y \): are material property.

Thus, \( \mu_d = \frac{\sigma_s}{\sigma_y} \) \( \rightarrow \) Amonton - Coulomb law.

At constant \( v \), time of contact reduces hence, \( \mu_d = \mu_d(v) \), decreases as \( v \) increases.

In velocity weakening regime, any fluctuation is unstable \( \rightarrow \) Stick-Slip. Stuck state is due to elastic loading and Slip state due to stress relaxation.

(a) \( M = 2.1 \text{ kg}, k = 1.5 \times 10^4 \text{ N m}^{-1}, \text{ and } V = 10 \mu \text{m s}^{-1} \) in paper–on–paper system. (b) Creep plot of the slider displacement \( x \text{ Vs } t \) for \( M = 0.32 \text{ kg}, k = 1.5 \times 10^4 \text{ N m}^{-1}, \text{ and } V = 5 \mu \text{m s}^{-1} \).
**Introduction:**

The crucial physical ingredient responsible for stick-slip behavior is “velocity-weakening” phenomena.

(a) Velocity-weakening law. A slight uncertainty in the stiffness leads to unstable motion i.e shown. (b) Schematic view of two rough surfaces. (c) Optical visualization of two rough epoxy resin blocks.
Friction process is complex:

- Adhesion, wear, interfacial layer, plastic deformation, smooth, roughness, contact electrification etc., are contribute to friction.

- Plastic deformation - contact radius, load - Hertz vs JKR, multiple scales in roughness
Electronic states deep in the gap of the polymer arising from side-groups. The location of the lowest occupied molecular orbital (LUMO) to highest occupied molecular orbital (HOMO) with respect to $E_F$ is shown.

Surface Force Apparatus (SFA) is a useful technique to study friction at these scales.

AFM, $R \sim \text{nm}$, $F_n \sim \mu\text{N-mN}$, $\sigma \sim \text{GPa}$;

SFA, $R \sim \mu\text{m- mm}$, $F_n = \text{a few mN}$, $\sigma \sim 50\text{MPa}$.

When a microcantilever with a nano-scale tip is scanned laterally over a surface to measure the nano-scale force it exhibits stick-slip motion.

The nature of stick-slip depends on probe stiffness, structure of the tip, surface energy and scan parameters (load, velocity, etc.)
Experiment:

- SFA experiment of Budakian and Putterman (Phys. Rev. Lett., 85, 1000 (2000)) - Friction ascribed to the formation of bonds arising from contact charging

- The cantilever of a SFA was dragged with a velocity of a few $\mu m/s \sim 10\mu m/s$ on polymethylmethacrylate (PMMA) surface.

- The $R$ of the gold ball 0.5 mm, $V$ a few $\mu m/sec \sim 10 \mu m/sec$.

- Typical measured charge density $\sim 10^8$ charges/mm$^2$.

Experimental Set-Up

Image of charge during stick-slip motion
Puttermann et al. (2000): main results to be explained: Frictional sliding is due to contact charging - bonds are formed. (1) correlation between force (stick-slip events) and charge transfer.

(2) the total force is proportional to the total charge deposited over a scan length. Scale factor $\alpha$.

(3) The value of $\alpha \sim 0.4 \text{ eV/Å}$; $\alpha$ constant for $68 \leq F_n \leq 106 \text{ mN}$.
- $\alpha$ same for smooth sliding,
- fewer stick-slip events of larger magnitude for higher $F_n$

(4) $\alpha \sim 0.4 \text{ eV/Å}$ is close to the energy window for transfer of charge from Fermi level
Why doubt their claims?

A) Force of Attraction due to dipole layer

\[ F_c = \frac{(\mu \pi a^2 \sigma)}{(2\epsilon_0 \kappa)} \]

Hertz radius is \( a_H^3 = 3F_n R/4E^*; \)

Using \( R = 0.5\text{mm}; F_n = 0.1\text{N}; E^* = 1.3\text{GPa}; \)
\[ a_H = 22\mu\text{m} \]

Using this and
\[ \sigma = 10^8 \text{esu/mm}^2 = 1.67 \times 10^{-5} \text{C/m}^2; \epsilon_0 = 8.85 \times 10^{-12} \text{C}^2/\text{Nm}^2; \kappa = 3.5 \]

It turns out \( F_c = 10^{-9} \text{N}, \) but \( \delta F \sim \text{mN} \)

B) Stress \( \tau = \frac{F_n}{(\pi(2.2)^2 \times 10^{-10})} = 6.9 \times 10^7 \)

\( \tau_y \) for PMMA is 10\( \text{MPa} \)

\( \tau_y \) for Gold is 80\( \text{MPa} \) Thus asperities are plastically deformed.
**Toy Model**

- Our aim is to build a simple model to explain the major features of the experiments.

- A toy for stick-slip and couple to charging and charge transfer equation.

\[
\ddot{X} = F - A \dot{X} - \frac{\dot{X}/v_m}{1 + (\dot{X}/v_m)^2} - f_{ch}\Sigma^2 \tag{1}
\]

\[
\dot{\Sigma} = \frac{1 - \Sigma}{\tau} - \frac{\dot{X}\Sigma}{a} \tag{2}
\]

\[
\dot{\Sigma}_d = \frac{\dot{X}\Sigma}{a} \tag{3}
\]

\[
\dot{F} = v_a - \dot{\dot{X}} \tag{4}
\]

- Equation for dimensionless displacement $X$ of the gold tip.

- $A \dot{X}$ dissipative term, III term the velocity weakening law that has maximum at $v_m$.

- The last term is frictional resistive force from electrostatic adhesion.
**Results:**

The parameters used in the results reported here are $v_a = 0.02, v_m = 0.01, \tau = 2.0, a = 1.57$ and $f_{ch} = 2.56 \times 10^{-7}$.

Figure 3: (a,b) Plots of force versus displacement for $A = 3.0$ and $7.0$ respectively, and (c, d) displacement and cumulative charge as a function of time corresponding to (a).
Results clearly displays the correlation between the displacement and the cumulative charge deposited on the PMMA surface and both has same slope.

Fewer stick - slip events for larger $F_n$

The established correlation between $X$ and $Σ_d$ also suggests that the authors interpretation that macroscopic friction arises from 'collective effects of bonds' formed due to charge transfer is not entirely warranted.
Full Model

- Include all relevant physical mechanisms:
  (a) Single asperity
  (b) JKR vs Hertz
  (c) Visco-elasticity
  (d) Plastic deformation
  (e) Adhesion due to contact charging
(a) Hertz relation for the area of contact - contact radius and depth

\[ A_n = \pi a^2 = \pi \left[\frac{3RF_n}{4E^*}\right]^{2/3} = \pi Rz \]  

For \( E^* = 2GPa; F_n = 0.1N; R = 0.5mm \quad a_H \sim 2.67 \times 10^{-5}m \)

(b) JKR radius -

\[ a^3 = \frac{3R}{4E^*} \left( F + 3\pi \gamma R + \sqrt{6\pi R \gamma F + (3\pi R \gamma)^2} \right) \]  

\( a_{JKR} \sim 3.4 \times 10^{-5} \) for \( \gamma = 0.1J/m^2 \) and since \( a_{JKR} \preceq a_H \), use \( a_H \)

(c) Visco-elastic effect \( G \longrightarrow G + \eta \dot{\varepsilon} \)

(d) Plasticity of interface asperities: \( \dot{\varepsilon}_{ve} \longrightarrow \dot{\varepsilon}_{ve} + \dot{\varepsilon}_{pl} \).

(e) Contact charging contribution - Using this contact area of charging, the attractive force is given by \( \pi Rz \sigma^2 / 2\varepsilon_0 \kappa \).

(f) For contact radius \( (a \sim 28\mu m \text{ for } F_n = 0.1N) \), the force of attraction due to charges is \( \sim 10^{-9}N \), but serrations \( \delta F \preceq mN \).
Full Model

Model describes tip coordinates $x$ and $z$ (depth). Our equations are of the general form

$$m \ddot{y} = F_a - F_r$$

(7)

where $y$ represents $x$ or $z$,

- $F_a \rightarrow$ applied force and $F_r \rightarrow$ material response

- In equilibrium, $F = F_r$.

- In dynamic conditions, $F \neq F_r$ due to visco-elastic and plastic deformation.

Contributions to force:
- Projected area in $x$- and $z$-directions
  $$A_x = A_{x,0} z^{3/2} \sim \frac{2}{3} \pi \sqrt{2R} z^{3/2}.$$  
  $$A_n = \pi a^2.$$  

- Frictional force at the contactting interface is $A_{x,0} \tau_0 z^{3/2}$, $\tau_0$ is shear strength

- Frictional resistance arising from contact electrification = $\mu A_n \sigma^2 / 2 \varepsilon_0 \kappa$.

- Visco-elastic creep contribution to the flow = $\eta || A_z \dot{x} / D : A(z) = A_{x,0} z^{3/2}$. 
Model

- Plastic strain rate $\dot{\varepsilon}_p = \dot{\varepsilon}_0 (\tau / \tau_y)^n$ - Plastic deformation is continuous beyond the linear visco-elastic flow.
  \[ = \eta_{\|} A_{x,0} z^{3/2} \frac{\dot{x}}{D} \left[ 1 - \left( \frac{F}{A_{x,0} \tau_0 z^{3/2}} \right)^n \right] \]

- Equation of motion along $x$- direction.
  \[ m\ddot{x} = F - F_r \]
  \[ m\ddot{x} = F - A_{x,0} \tau_0 z^{3/2} - \mu \pi R z \frac{\sigma^2}{2\kappa \varepsilon_0} \]
  \[ - \eta_{\|} A(z) \frac{\dot{x}}{D} \left[ 1 - \left( \frac{F}{A_{x,0} \tau_0 z^{3/2} + \mu \pi R z \sigma^2 / 2\kappa \varepsilon_0} \right)^n \right], \]  \[ \dot{F} = K_{\|} (V_a - \dot{x}), \]
Model

The equation of motion along $z$– direction is

$$
\begin{align*}
    m\ddot{z} &= F_n - \frac{4}{3} R^{1/2} z^{3/2} \left[ E^* + \frac{\eta}{D} \dot{z} \left( 1 - \left( \frac{F_n}{\tau_{y,n} \pi R z} \right)^q \right) \right] + \frac{c\eta}{D} \dot{x} - F \frac{z^{1/2}}{\sqrt{2R}} \tag{11}
\end{align*}
$$

The elastic response $= \frac{4}{3} R^{1/2} z^{3/2} E^*$. 

$\eta/\dot{z}/D =$ visco-elastic creep along $F_n$. 

Plastic strain rate $= -\frac{\eta}{D} \dot{z} \left( \frac{F_n}{\tau_{y,n} \pi R z} \right)^q$. $q$ exponent .

The tip creeps in the $z$ direction, the position of tip center $x$, also creep along $x$. Additional creep contribution $c\eta/\dot{z}/D$.

Note Eq. (8) gives $a^2 = \left[ 3RF_n / 4E^* \right]^{2/3}$ Hertz radius.
Model

The evolution of the charge density $\sigma$ is given by

$$\dot{\sigma} = \frac{\sigma_m}{t_a} \left(1 - \frac{\sigma}{\sigma_m}\right) - \frac{\dot{x}}{D} \sigma$$

(12)

$\sigma_m$ = maximum charge density.

$t_a$ = time constant.

Total charge $\sigma_t = \pi R z \sigma$,

$$\dot{\sigma}_t = \frac{(\pi R z \sigma_m - \sigma_t)}{t_a} + \sigma_t \frac{\dot{z}}{z} - \frac{\dot{x}}{D} \sigma_t$$

(13)

The Scaled Equations

$D = F_{max}/K_{||}$ and a time scale determined by $\omega^2 = K_{||}/m$.

The scaled variables are: $x = XD$, $z = ZD$, $F^s = F/K_{||} D$, $\tau_0 = \tau_0 A_{x,0} D^{1/2}/K_{||}$,

$F_n = F_n/K_{||} D$, $K_\perp = \frac{4}{3}(RD)^{1/2} E^*$, $\eta_\perp = \frac{4}{3}(RD)^{1/2} \omega \eta_\perp / K_{||}$, $\eta_\parallel = A_0 \eta_\parallel \omega / K_{||} D$,

$\tau_y = \tau_{y,n} \pi R / K_{||}$, $v_a = V_a / \omega D$, and $T_a = t_a \omega$.

$\Sigma = \pi RDZ \sigma / \sigma_0$ with $\sigma_0 = (2\pi k \epsilon_0 RD^2 K_{||})^{1/2}$ and $\Sigma_m = \pi RD \sigma_m / \sigma_0$, 
The Scaled Equations

\[ \ddot{X} = F^s - \bar{\tau}_0 Z^{3/2} - \mu \frac{\Sigma^2}{Z} - \bar{\eta}_\parallel \dot{X} \left[ 1 - \left( \frac{F^s}{\bar{\tau}_0 Z^{3/2} + \mu \frac{\Sigma^2}{Z}} \right)^n \right], \tag{14} \]

\[ \ddot{Z} = \bar{F}_n - \left[ \frac{K_\perp}{K_\parallel} + \bar{\eta}_\perp \dot{Z} \left( 1 - \left( \frac{\bar{F}_n K_\parallel}{\bar{\tau}_y Z} \right)^q \right) \right] + c\bar{\eta}_\perp \dot{X} \right] Z^{3/2} - \beta F^s Z^{1/2}, \tag{15} \]

\[ \dot{\Sigma} = \frac{\Sigma_m Z - \Sigma}{T_a} - \dot{X} \Sigma + \frac{\dot{Z}}{Z} \Sigma, \tag{16} \]

\[ \dot{F}^s = v_a - \dot{X}. \tag{17} \]

- The scaled equation for charge transferred (to the substrate) is \( \dot{\Sigma}_d = \dot{X} \Sigma. \)
- Stick-slip due to feedback loop.
- Steady state unstable - Instead of stability analysis, calculate force in stationary condition

\[ F^s = \frac{\bar{\tau}_0 \bar{F}_n}{\left[ \frac{K_\perp}{K_\parallel} + c\bar{\eta}_\perp v_a \right]} \tag{18} \]
### Parameters

<table>
<thead>
<tr>
<th>$R$</th>
<th>$m$</th>
<th>$K_\parallel$</th>
<th>$F_n$</th>
<th>$V_a$</th>
<th>$\sigma_m$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(mm)</td>
<td>(kg)</td>
<td>(N/m)</td>
<td>(mN)</td>
<td>(μm/s)</td>
<td>(C/m$^2$)</td>
</tr>
<tr>
<td>0.5</td>
<td>$10^{-5}$</td>
<td>47</td>
<td>$68 - 106$</td>
<td>$\sim 10$</td>
<td>$1.67 \times 10^{-5}$</td>
</tr>
<tr>
<td>$E^*$</td>
<td>$\eta_\parallel$</td>
<td>$\eta_\perp$</td>
<td>$\tau_0$</td>
<td>$\tau_{y,n}$</td>
<td>$\sigma_0$</td>
</tr>
<tr>
<td>GPa</td>
<td>Pa.s</td>
<td>Pa.s</td>
<td>MPa</td>
<td>MPa</td>
<td>(nC/m$^2$)</td>
</tr>
<tr>
<td>1 − 3</td>
<td>...</td>
<td>...</td>
<td>0.1 − 10</td>
<td>1 − 50</td>
<td>3.0</td>
</tr>
<tr>
<td>$K_\perp$</td>
<td>$\bar{\eta}_\parallel$</td>
<td>$\bar{\eta}_\perp$</td>
<td>$\bar{\tau}_0$</td>
<td>$\bar{\tau}_y$</td>
<td>$\Sigma_m$</td>
</tr>
<tr>
<td>$\sim 10^6$</td>
<td>$10^2$</td>
<td>$8 \times 10^8$</td>
<td>1.0</td>
<td>2500</td>
<td>0.0167</td>
</tr>
</tbody>
</table>

- The magnitude of $\tau_0 \sim \mu \tau_s$, where $\tau_s$ is the shear yield stress and $\mu \sim 0.3$ is the friction coefficient.
- Other parameters used are: $A_0 \sim a^2 \sim 10^{-10} m$, $\kappa = 3.3$, $v_a = 1.45 \times 10^{-5}$, $T_a = 2$ and $c = 0.122$. Choosing $F_{max} = 47 \text{ mN}$ gives $D = 10^{-3} m$ and thus the range of $\bar{F}_n$ corresponding to $68 - 106 \text{ mN}$ is $1.446 - 2.26$. 
Results

\[ F_n = 2.0, \]

\[ 3.09 \times 10^4 < \alpha_s < 4.18 \times 10^4 \text{ for } 1.446 < F_n < 2.26. \]

\[ F_n = 3.5. \]

\[ F_n = 2.0 \text{ and } \Sigma_m = 0.835 \left( \sigma_m = 5 \times 10^{-4} C/m^2 \right) \]
Correlation between stick-slip events and charge transfer.

The ratio of maximum deviation ($1.09 \times 10^4$) to the mean ($3.365 \times 10^4$) of $\alpha$ is $\sim 30\%$.

- Comparable with $25\%$ maximum scatter for the range of $F_n = 68 - 106 \text{ mN}$ in expt.

Thus, $\alpha^s$ independent of load is due to limited range of $F_n$, not due to true independence. $A_n \propto F_n^{2/3}$, $\alpha$ depends on load.
Results

$\alpha^s$ same for smooth sliding, steady state.

Note $A_n \rightarrow 1$ to 1.34, $F_n \rightarrow 68$ to 106 mN. Not really independent but small range of $F_n$. 
Results

- For larger loads fewer events of larger magnitude (on an average) for the same scan length.

\[ \int K \dot{x} dt = \alpha \int \dot{\sigma} dt = \alpha \int (\dot{x} \sigma_t / D) dt \longrightarrow K \parallel D \simeq \alpha \pi R \sigma_{asy} \sigma_{asy} \]

Using \( z_{asy} \) and \( \sigma_m \) leads to \( \alpha \sim 50 \text{ eV/Å} \) for \( F_n = 94 \text{ mN} \) which is even after discounting the ideal nature of the model.

- Possible reasons: Experimental charging area large; Roughness

- In reality in experiments, charging radius \( R_c \) is 10-20 times \( a \) for

\( \sigma \sim 1.6 - 2.4 \times 10^8 \text{ charges/mm}^2 \).

Thus, \( A_c = \beta A_n \) with \( \beta \sim \left( \frac{R_c}{a} \right)^2 \sim 100 - 400 \) or use higher charge density.

- Using \( \beta A_n \) in place of \( A_n \), we get \( \alpha \sim 0.5 - 0.125 \text{ eV/Å} \) that is consistent with the reported value.

- Roughness can also be contribute to - typically - 10%

- Static friction is less than sliding friction.
Summary and Conclusions

- The correlation between the stick-slip events and charge transfer reproduced.

- Lack of dependence of the scale factor $\alpha$ on normal load is due to the small range of values used in expt. Indeed, the scale factor is inversely proportional to the area of contact. Using $A_c = \pi a^2$ gives

Using experimental contact area $A_\varepsilon = \beta A_c$, $\beta \sim 100 - 400$; give $\alpha = 0.25 - 0.5$ eV.

- These features follow naturally due to the separation of time scales of stick and slip events.

- As most experimental features are captured, the model provides an alternate explanation for the results.

- Stick-slip events in our model are not influenced by charge.

- Plastic deformation of the interfacial layer is responsible for stick-slip dynamics that arises due to a competition between the internal relaxation time scales (visco-elastic and plastic deformation time scales etc) and that due to the pull speed.
THANK YOU